



Short communication

A review of the performance and analysis of proton exchange membrane fuel cell membrane electrode assemblies

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H I G H L I G H T S

- The performance of the CCMs method is approximately 18% better than that obtained by using GDEs.
- The performance of the self-fabricated membrane was similar to that of NRE212.
- The self-humidifying membrane operated over more than 300 h while it was supplied with dry hydrogen and oxygen.
- A matched 250 μm silicon gasket with different assembly torques of 5–30 Kgf cm can get similar performances.

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This study describes a performance review of several membrane electrode assemblies (MEAs) for proton exchange membrane fuel cells (PEMFC). First, different methods for preparing catalyst-coated membranes (CCMs) and gas diffusion electrodes (GDEs) are presented to show that the power density of the CCMs method is approximately 18% better than that obtained by using GDEs. Second, different thickness membranes and a self-fabricated membrane are discussed. The self-fabricated membrane used a PTFE microporous membrane as a backing structure and was impregnated with Nafion for reinforcement. Third, we compared the performance differences of four different amounts of platinum loaded in Nafion-bonded CCMs to prove that more platinum loading can produce better performance linearly at a platinum loading from 0.1 to 0.4 mg cm^{-2} . Fourth, a water storage zone was created on the surface of the GDL. The voltage–time curve shows that the voltage is maintained at $0.650 \text{ V} \pm 0.015 \text{ V}$ over more than 300 h while supplying with dry hydrogen and oxygen. Fifth, a matched 250 μm silicon gasket was used to test the performance of a standard MEA with different torques of 5–30 Kgf cm, and the performance differences are within 10%.

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1. Introduction

During the past two decades, the proton exchange membrane fuel cell (PEMFC) has been regarded as one of the most promising alternative power sources due to its high efficiency and low emissions [1–6]. Fuel cells convert the chemical energy present in hydrogen and oxygen into electricity, generating only heat and water. In theory, the PEMFC is the cleanest renewable energy source, and its fuels (hydrogen and oxygen) are abundant on earth. Recently, great progress has been achieved in improving the power density, operational stability and stack design of PEMFCs. However, there are still many challenges that need to be overcome for profitable commercial application. Improving the spraying method,

lowering the Pt loading, recycling the liquid water generated at the cathode, and simplifying the cell manufacturing process are all ways to bring PEMFCs toward commercial reality [7,8].

Membrane electrode assemblies (MEAs) are the most important parts of PEMFCs. A higher performance MEA could be obtained by improving the contact between the electrolyte and catalyst layers [9,10], and such a MEA must be designed as simply as possible to meet weight and volume requirements [11–13]. Coating the catalyst layer onto a Nafion membrane surface, referred to as a catalyst-coated membrane (CCM), has several advantages, such as lower Pt loading with higher performance [14,15]. Nafion, used as the solid polymer electrolyte in PEMFCs, is the most common and available product. However, different solid polymer electrolytes have been developed, such as sulfonated FEP–Nafion hybrid membranes [16], PFSA–ePTFE composite membranes [17], and membranes utilizing the addition of microporous materials together with Nafion for improved strength [18,19].

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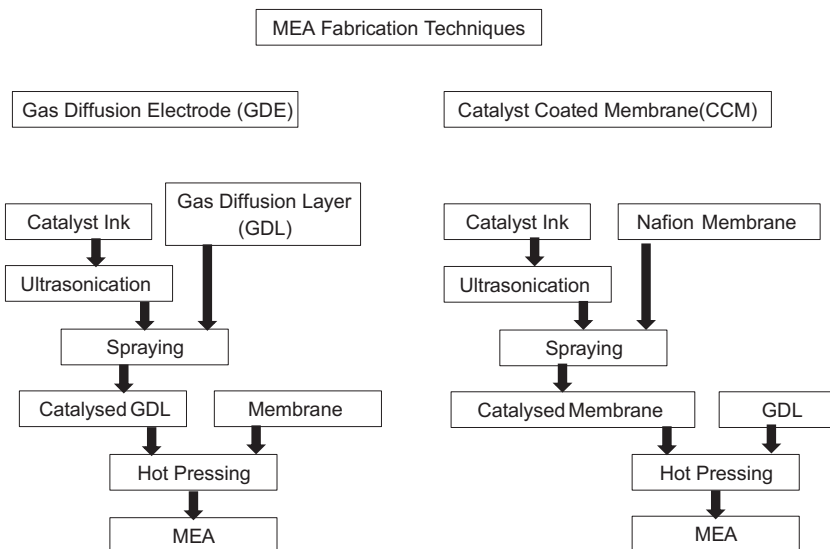


Fig. 1. Membrane electrode assembly fabrication techniques.

To reduce humidifier consumption, substantial efforts have been made to develop self-humidifying MEAs. Most of the ideas have focused on incorporating highly dispersed nanosized Pt particles in the Nafion membrane [20–23] or modifying an area of the gas diffusion layer to capture water [24,25]. Satisfactory proton conductivity has been achieved with a self-humidifying MEA [21,26]. Dispersing nanosized Pt particles in the Nafion membrane would allow these Pt nanoparticles to catalyze the recombination of hydrogen and oxygen diffusing into the Nafion membrane. Moreover, these Pt nanoparticles could decrease reactant cross-over through the Nafion membrane and increase its water content, reducing proton transport resistance. In this way, a few disadvantages, including the high cost and difficult formation of an electron-conducting path via the network of dispersed Pt or Pt/C nanoparticles, are discussed [27].

This study presents numerous MEAs, including those produced by spray-based methods (CCMs and GDEs) and processing different Pt loadings ($0.1\text{--}0.4\text{ mg cm}^{-2}$), MEAs consisting of a reinforced self-fabricated membrane (compared to NRE-211 and NRE-212), a self-humidifying membrane (at more than 300 h operation), and a tested combination of 250 μm thickness of silicon gasket and Sigracet 10BC GDL are presented. This study primarily analyzes and reviews the performance of several MEAs, which are characterized by changing or processing partial layers, such as the membrane, catalyst layer, and gas diffusion layer. Many components of MEAs are still being developed and optimized for commercial PEMFC products.

2. Experimental

2.1. Materials

All materials were used in this study without any further treatment, as-received. Dupont Nafion® NRE-211 (25 μm) and NRE-212 (50 μm) membranes and Nafion® solution–DE-520 were purchased from Dupont's Taiwan agent: Tension Energy Inc. Platinum, nominally 50%, on carbon black (50 wt% Pt/C Johnson Matthey HiSpec™) was purchased from Alfa-Aesar® mixed as catalyst ink. Only one type of commercial GDL was used in the study, Sigracet SGL 10BC (415 μm), which exhibited the best performance for PEMFC [28], and a silicon gasket (250 μm) was

chosen to match the compressed thickness of the GDL in the unit cell combination.

2.2. MEA fabrication and testing

The procedures for MEA fabrication are given in Fig. 1. There are several methods that can be used to coat catalyst ink on the surface of a membrane or GDL, such as ultrasonic spraying, air spraying, brush coating, and the transfer of ink from a PTFE sheet. In this study, the air spraying method was used to achieve high quality MEAs. Catalyst ink was prepared by using 50 wt% of Pt/C instead of 20 wt%. There are two main reasons for this. First, 20 wt% Pt/C is difficult to disperse well if we set the expected Pt loading to 0.4 mg cm^{-2} . The catalyst ink would need to be dissolved in larger solution volume, such as isopropyl alcohol. Second, to achieve the desired Pt loading, spraying 20 wt% Pt/C catalyst ink takes more time than that does using 50 wt% Pt/C.

Catalyst inks were prepared by ultrasonic mixing (45 kHz ultrasonic bath, 200 W max ultrasonic power, 30 min) of the commercial 50 wt% Pt/C, Nafion solution (5 wt%), an appropriate amount of isopropyl alcohol, and a few ml of deionized water. Deionized water needs to be added first to moisturize the 50 wt%

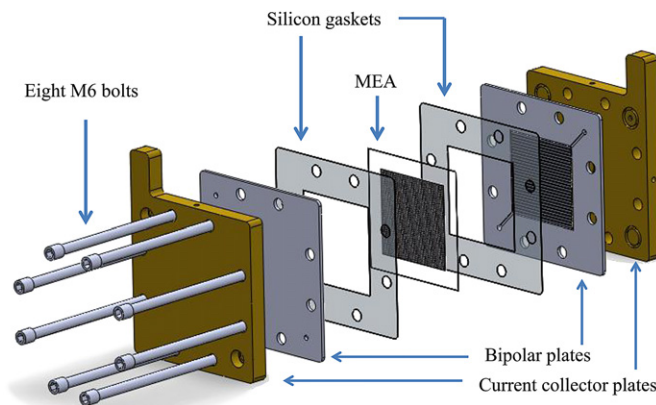


Fig. 2. CAD drawing of the unit cell.

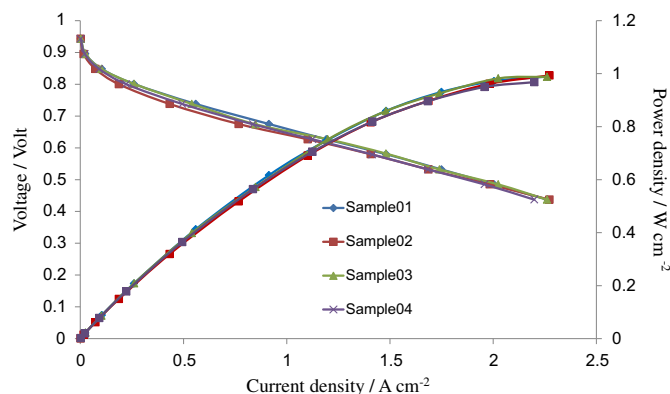


Fig. 3. Polarization and power density curves of 4 random MEAs coated with GDEs method at a platinum loading: 0.4 mg cm^{-2} at the anode and cathode.

Pt/C, which would react and burn when it contacts the Nafion solution or isopropyl alcohol if it is dry. The ratio of 50 wt% Pt/C to Nafion was typically 25%–75% by weight. Before spraying, the membranes or GDLs were put on a stainless-steel-coated chromium plate and heated to approximately 90°C . Heat transmits well over the plate, and this process helps to vaporize unnecessary solution. The catalyst inks were air-sprayed onto the membranes or GDLs at various catalyst loadings from 0.1 to 0.4 mg cm^{-2} . The active area of all MEAs was 25 cm^2 . The last step of the MEA fabrication was completed by hot-pressing at 140°C for 120 s under a pressure of 2.0 MPa.

The proton exchange membrane fuel cell tests were all assembled in a single cell [Fig. 2], serpentine bipolar plate channels and operated at 70°C and 100% relative humidity, with ambient pressure on both the anode and cathode sides. The fuels given were H_2 and O_2 (1.5 and 2.0 stoichiometries, respectively). In an exceptional test, the lifetime test of the self-humidifying MEA provided with dry hydrogen and oxygen (1.5 and 2.0 stoichiometries), the cell was not heated from the outside. The temperature increased due solely to the heat that was self-generated. All polarization curves were measured until the current of each MEA was stable at constant voltage activation (0.6 V).

2.3. Self-fabricated membrane and pre treatment

First, a $10 \text{ cm} \times 10 \text{ cm}$ PTFE microporous membrane ($25 \mu\text{m}$ thick, pore size 50 nm, Shanghai Dagong New Material Co., Ltd.) was placed on the stainless steel coated chromium plate and heated

to approximately 90°C . Approximate 3 ml of Nafion solution was added dropwise to impregnate the whole PTFE microporous membrane, until unnecessary solution was vaporized. The PTFE microporous membrane was white before Nafion solution was added. After Nafion addition, it became transparent, similar to the NRE-211 or NRE-212. The Nafion impregnation procedure required a few repetitions for completion; it was ensured that all pores of the PTFE microporous membrane were filled with Nafion. Before coating with catalyst inks, the Nafion-filled PTFE microporous membrane was put into an oven to anneal at 140°C for 1 h. Then, the self-fabricated membrane was cleaned to remove the remaining organic and inorganic contaminants [29,30] and then converted into H^+ form for use.

After annealing in the oven, the self-fabricated membrane demonstrated brown coloration, and it was first immersed in boiling 5 wt% H_2O_2 for 1 h to remove organic and inorganic contaminants. After the membrane was washed with deionized several times, it was boiled in $0.5 \text{ mol l}^{-1} \text{H}_2\text{SO}_4$ solution for 1 h for conversion to its H^+ form. Then, the self-fabricated membrane was rinsed in boiling deionized water at least twice to remove the sulfuric acid completely and stored in pure water for later coating of catalyst inks. The procedures for the CCM and hot pressing of GDLs were the same as described above. The Pt loading for both sides was 0.4 mg cm^{-2} .

2.4. Self-humidifying membrane

The water storage area was processed on the surface of a commercial GDL that has two layers: carbon fibers (consisting of 30 wt% PTFE) and a microporous layer coated with 20 wt% PTFE containing carbon black (XC-72). For the anode and cathode, the ratio of XC-72 carbon black to Nafion solution was 3:1 by weight, and 1.0 mg cm^{-2} loading and coating on the surface of a microporous layer was observed after ultrasonic mixing. The self-humidifying MEA was prepared by hot-pressing with the CCM (Pt loading for the anode and the cathode: 0.4 mg cm^{-2}) at 140°C for 120 s under a pressure of 2.0 MPa.

3. Results and discussion

3.1. Performance of replicate samples

To distinguish systematic pattern from a chance or random variation, we randomly chose 4 MEAs from 25 samples at a platinum loading of 0.4 mg cm^{-2} to test and clarified that our process and air spraying skills can show excellent uniformity. In Fig. 3

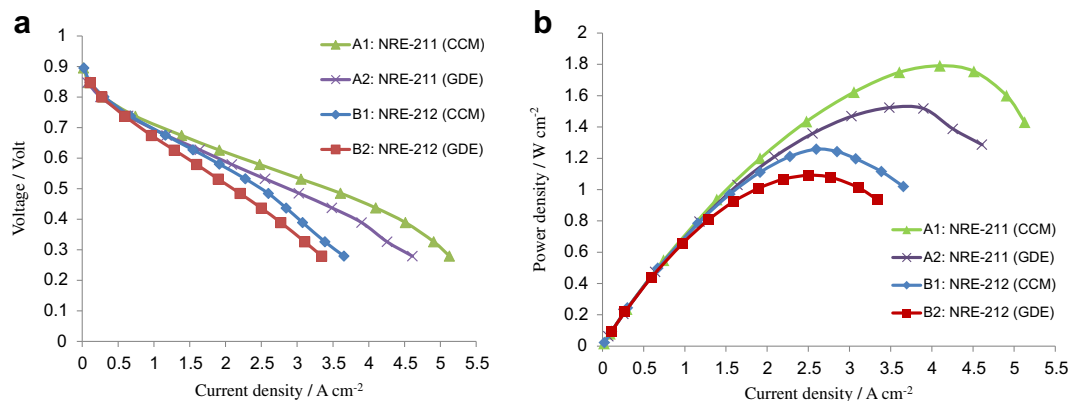


Fig. 4. (a) Polarization curves: air-sprayed CCMs and GDEs (NRE-211, NRE-212) with anode and cathode loadings of 0.4 mg cm^{-2} . (b) Power density curves. The peak power density was 1.791 W cm^{-2} at 0.45 V for CCM (NRE-211).

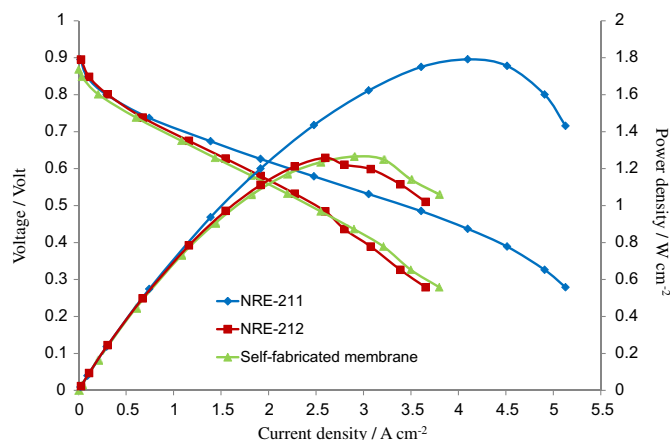


Fig. 5. The polarization and power density curves of NRE-211, NRE-212, and self-fabricated membrane with Pt loading 0.4 mg cm^{-2} at the anode and cathode.

shows the polarization curves of four MEAs coated with GDEs method. The peak power densities are 989, 993, 987, and 967 W cm^{-2} operated at 65°C and 100% relative humidity with H_2 and O_2 , means that the uniformity of each MEA is within 3%.

3.2. Performance comparison of CCM and GDE

Four MEAs were tested for their performance. A1: NRE-211 (CCM); A2: NRE-211 (GDE); B1: NRE-212 (CCM); B2: NRE-212 (GDE). The Pt loading for both sides was 0.4 mg cm^{-2} . Fig. 4 shows the polarization (a) and power density (b) curves for MEAs using different coating methods and membrane thicknesses. It can be observed from Fig. 4(b) that A1 exhibits the best performance with a peak power density 1.791 W cm^{-2} , of A2 is 1.523 W cm^{-2} , of B1 is 1.258 W cm^{-2} , and of B2 is 1.090 W cm^{-2} . According to the polarization curves, regardless of whether NRE-211 or NRE-212 was used, the peak power density of the CCMs is approximate 18% better than that of the GDEs.

When CCMs and GDEs are compared, the performance of CCMs is absolutely better than GDEs in theory and in practice because the catalyst inks are all sprayed on the surface of the membrane. For GDEs, even though the microporous layer is formed by nanosized carbon black, partial catalysts will permeate into the carbon fiber structures when spraying and become waste catalysts. Consequently, we can say that a greater percentage of platinum reacts with hydrogen and oxygen on the CCMs than it does on the GDEs. However, MEAs can be fabricated more quickly with GDEs than

with CCMs because the membranes in CCMs undergo swelling and wrinkling during the spraying deposition of the catalyst layers.

3.3. Performance comparison of NRE211, NRE212 and the self-fabricated membrane

The irregular thickness of the self-fabricated membrane is between $30 \mu\text{m}$ and $48 \mu\text{m}$, and it is coated with a Pt loading 0.4 mg cm^{-2} . Fig. 5 shows the polarization curves for NRE-211, NRE-212, and the self-fabricated membrane. Predictably, it can be observed that the performance of the MEA increases as the membrane is thinned. However, in comparing the self-fabricated membrane to NRE-212, a direct relationship between the increase in MEA performance and the membrane thickness is not obvious because of the rough surface. The thin membrane has a good proton transfer rate because the gap between the anode and cathode is short. The self-fabricated membrane is thinner than NRE-212 on average; it is a hand-made laboratory sample compared with NRE-212, which is a commercial product. There is no doubt about the quality of NRE-212.

Furthermore, a PTFE microporous membrane can be used as a backing structure to reinforce the self-fabricated membrane. DuPont membranes are easily torn apart once there is a crack in them, especially thin membranes such as NRE-211 ($25 \mu\text{m}$). The membrane thickness is a main factor by which the maximum performance of a MEA can be determined if the catalyst spraying technique is mature and consistent. While a thinner membrane can obtain better performance, it is not easy to spray catalyst layers during the MEA fabrication procedure.

3.4. Performance comparison of Pt loading (0.1, 0.2, 0.3, 0.4 mg cm^{-2})

The performance of CCMs of various loading was investigated on a Sigracet 10BC GDL. A comparison of the polarization (a) and power (b) curves for various platinum loadings of 0.1, 0.2, 0.3, and 0.4 mg cm^{-2} is shown in Fig. 6. The performance of the MEAs predictably increased when the platinum loading was increased. The peak power densities for the 0.1, 0.2, 0.3, and 0.4 mg cm^{-2} loadings are 0.943, 1.091, 1.167, and 1.258 W cm^{-2} , respectively, as shown in Fig. 6(b).

However, the performance at a loading of 0.4 mg cm^{-2} is not twice that observed at a loading of 0.2 mg cm^{-2} . In Fig. 7, the linear relationship between the maximum power density and the catalyst loading with a slope of almost 1, means that the two are directly proportional, the maximum power density is a strong function of the catalyst loading at platinum loadings from 0.1 to 0.4 mg cm^{-2} .

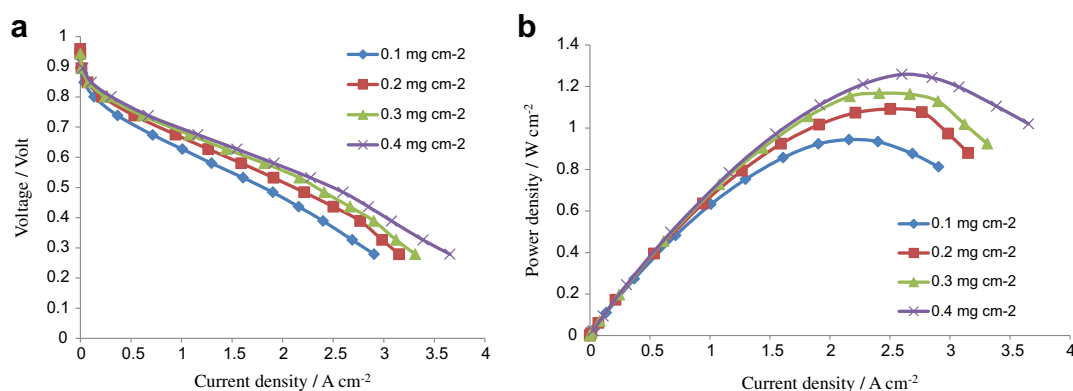


Fig. 6. The polarization (a) and power density (b) curves of CCMs with different Pt loadings: 0.1, 0.2, 0.3, and 0.4 mg cm^{-2} at the anode and cathode.

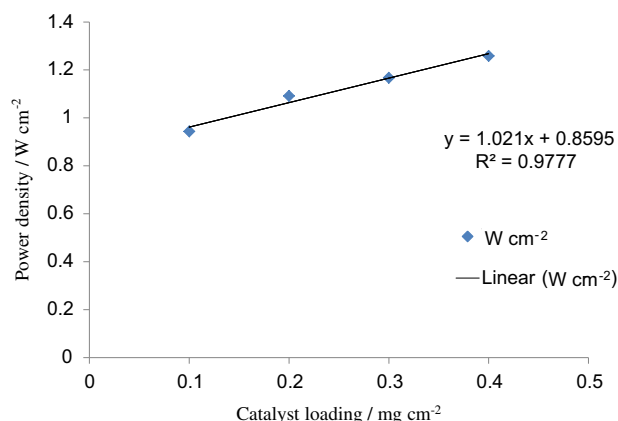


Fig. 7. The linear relationship between the max power density and the catalyst loading.

Consequently, extremely high performance cannot easily be obtained by adding more platinum. For commercial applications, the cost and performance value need to balance, or resources are wasted. Moreover, there is a mass transport issue at a loading of 0.4 mg cm^{-2} compared to the 0.1 mg cm^{-2} loading. Millington et al. [31] give a highlighted Nyquist plot shown that the thickness at a high loading is much higher in turns leading to a higher mass transport resistance.

3.5. Performance and lifetime of self-humidifying membrane

Extra water storage area was sprayed on the surface of a 10BC GDL. Before the lifetime test without any humidity supplied for hydrogen and oxygen, the self-humidifying MEA was activated at constant voltage (0.6 V) until the current density was stable and initially provided with fuels at 100% relative humidity at the anode and cathode to moisturize the membrane. Supplying fuels from the lower inlets of both sides is a trick to keep water inside of the unit cell as long as possible, especially the cathode side. Water is generated at the cathode side and accumulates at the bottom of the flow-filled plate. Oxygen is supplied from the lower inlet, which resembles a small bubble-type humidifier inside the unit cell to extend moisture content.

Fig. 8(a) shows the lifetime of the self-humidifying membrane with dry hydrogen and oxygen. The measurement was performed at constant current (1 A cm^{-2}) and heat generated by the PEMFC heated the cell to 55°C automatically. Under these operation conditions, the voltage was maintained at $0.650 \text{ V} \pm 0.015 \text{ V}$, and the device lasted for more than 300 h. In Fig. 8(b), two polarization curves show the performance of the fresh self-humidifying membrane and the same one after 300 h. If a proton exchange membrane is depleted of water, the proton transfer rate will decrease and cause irreversible damage to the membrane. Although the self-humidifying membrane is provided with dry hydrogen and oxygen, the maximum power density decays approximately 7% after 300 h (fresh: 1.095 W cm^{-2} , after 300 h: 1.017 W cm^{-2}), which shows that the function of the water storage area is successful for extended times.

3.6. Performance of assembly under different torques (5, 10, 15, 20, 25, 30 Kgf cm)

According to the official specification of the Sigracet 10BC GDL [32], the 10BC exhibits better performance due to its 3D non-woven structure [31]. For that reason, the 10BC has an open pore structure that leads to higher air permeability and is more compressible and resilient. We measured the hot-pressed MEA which the total thickness was approximately $660 \mu\text{m}$ that consists of two GDLs, two catalysts layers ($30 \mu\text{m}$ each in theory), and a membrane ($50 \mu\text{m}$). After the hot-pressing procedure (at 140°C for 120 s under a pressure of 2.0 MPa), the thickness of the 10BC is reduced from $415 \mu\text{m}$ to approximately between $280 \mu\text{m}$ and $310 \mu\text{m}$. A $250 \mu\text{m}$ thick silicon gasket was chosen to sit around the edge of the GDL and then assembled with bipolar plates and current collector plates (Fig. 2). The main purposes were to seal the PEMFC to prevent hydrogen and oxygen from leakage and to adjust the compression of GDLs.

Fig. 9(a) shows that the stressed thickness of one silicon gasket with given torques from 5 to 30 Kgf cm, without a membrane and GDL. After several tries, the thickness of $250 \mu\text{m}$ and the hardness of 50° shore A is the most suitable silicon gasket for Sigracet 10BC GDL. The thickness of the silicon gasket does not change significantly after 15 Kgf cm, which means that the compression of the GDL is similar at torques of 15–30 Kgf cm. From torques of 15 to 30 Kgf cm, the thickness of the silicon gasket is from 0.185 mm to 0.170 mm. In Fig. 9(b), the polarization curves show the performance of different torques of the same MEA. Initially, the PEMFC is

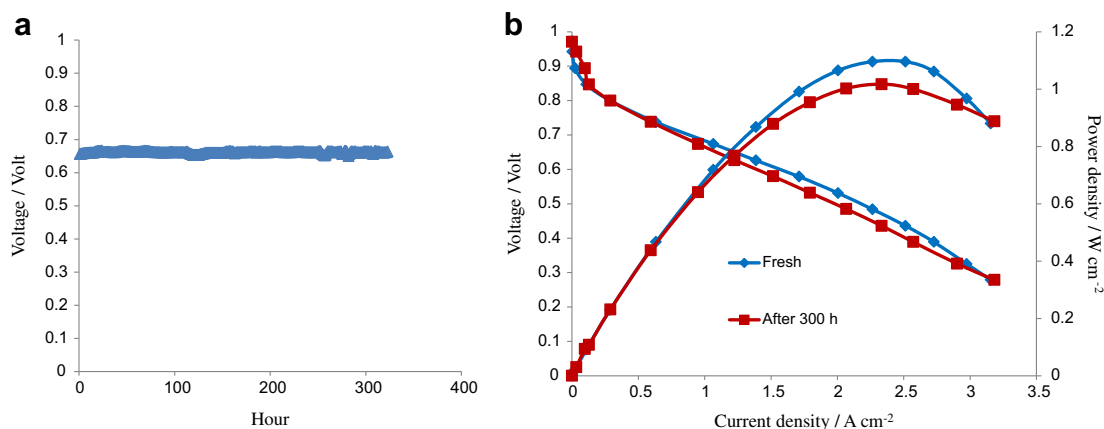


Fig. 8. (a) The voltage–time curve shows that the voltage of the self-humidifying membrane was stable during 300 h of operation. The test conditions were at constant current (1 A cm^{-2}), self-heated at approximate 55°C , and provided with dry hydrogen and oxygen. (b) The polarization curves show the performance of the fresh self-fabricated membrane and the same one after 300 h of operation.

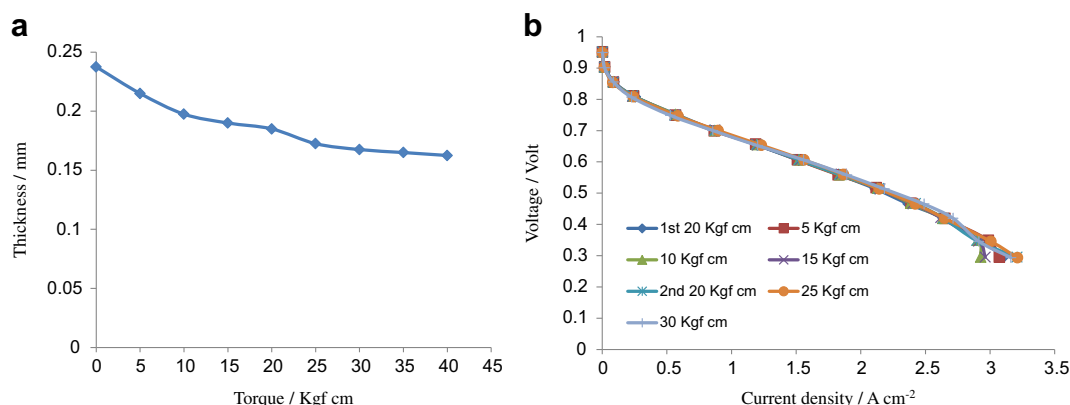


Fig. 9. (a) The compression of a 250 μm thick silicon gasket. (b) The polarization curves of a MEA tested at different torques from 5 to 30 Kg/cm.

assembled with 20 Kg/cm, and the first I – V curve is taken after activation. Then, the PEMFC is loosened to a torque of 5 Kg/cm and the I – V curve is measured again. The steps are performed several times by tightening the unit cell 5 Kg/cm each time and recording performance changes. Fig. 9(b) shows that the performances are similar, even if the torque is different, which means that the assembly torque is not a performance issue when a correct silicon gasket is used to match the GDL thickness. Thus, in this study, an excellent combination is obtained when a 250 μm thick silicon gasket is assembled with a Sigracet 10BC GDL after hot pressing at 140 $^{\circ}\text{C}$ for 120 s under a pressure of 2.0 MPa. The differences in performance are within 10% (@0.6 V).

4. Conclusion

In this paper, we reported a review of membrane electrode assembly (MEA) fabrication for PEMFC by replacing or processing different parts, such as the membrane, catalyst layer, and gas diffusion layer. We have reported that MEAs were fabricated with catalyst-coated membranes (CCMs) and gas diffusion electrodes (GDEs), and we have compared the performance between the two methods and with different membrane thicknesses. Normally, the performance of CCMs is approximately 18% better than that of GDEs. This paper also showed that the performance did not double if the platinum loading was doubled. For commercial products, the cost and performance values should be balanced. In addition, a reinforced self-fabricated membrane was compared to commercial membranes, and the self-fabricate membrane exhibited similar performance to NRE-212. Then, a water storage area created to capture water generated by the cathode operated successfully for more than 300 h without any humidity at either side. Finally, a suitable silicon gasket (250 μm thick) for a Sigracet 10BC GDL was tested at different assembly torques. Through the investigation of different torques, we found that the performance was nearly the same from 5 to 30 Kg/cm, with differences less than 10% (@0.6 V).

Acknowledgments

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